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PATENT AND TRADEMARK OFFICE

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P21844

TRANSMITTAL LETTER TO THE UNITED STATES  
DESIGNATED/ELECTED OFFICE (DO/EO/US)  
CONCERNING A FILING UNDER 35 U.S.C. 371

U.S. APPLICATION NO. (If known, see 37 CFR 1.5)

09/926826  
09/926826

INTERNATIONAL APPLICATION NO.

PCT/JP00/04004

INTERNATIONAL FILING DATE

19 June 2000

PRIORITY DATE CLAIMED

09 July 1999

TITLE OF INVENTION

FLAME RETARDANTS FOR RESINS AND FLAME-RETARDED RESIN COMPOSITIONS CONTAINING THE SAME

APPLICANT(S) FOR DO/EO/US

Noriaki TOKUYASU and Katumi KAMEDA

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information.

1. ☒ X. This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ X. This is an express request to promptly begin national examination procedures (35 U.S.C. 371(f)).
4. ☒ X. The US has been elected by the expiration of 19 months from the priority date (PCT Article 31).
5. ☒ X. A copy of the International Application as filed (35 U.S.C. 371(c)(2))
  - a. ☒ X. is attached hereto (required only if not communicated by the International Bureau).
  - b. ☐ has been communicated by the International Bureau.
  - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ X. An English language translation of the International Application as filed (35 U.S.C. 371 (c)(2)).
7. ☒ X. Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
  - a. ☐ are attached hereto (required only if not communicated by the International Bureau).
  - b. ☐ have not been communicated by the International Bureau.
  - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
  - d. ☐ have not been made and will not be made.
8. ☒ X. An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3))
9. ☒ X. An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☒ X. An English language translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (U.S.C. 371(c)(5)).

Items 11 to 16 below concern other document(s) or information included:

11. Assignee: DAIHACHI CHEMICAL INDUSTRY CO., LTD. of Osaka, JAPAN
12. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
13. ☒ X. An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
14. ☒ X. A FIRST preliminary amendment.  
☐ A SECOND or SUBSEQUENT preliminary amendment.
15. ☐ A substitute specification.
16. ☐ A change of power of attorney and/or address letter.
17. ☐ Figure of Drawing to be published
18. ☒ X. Other items or information:  
Cover Sheet and International Application as published in Japanese.  
PCT/RO/101-PCT Request (in Japanese).  
PCT/IPEA/409 (in English and Japanese).  
PCT/IB/338.  
PCT/ISA/210 (in English and Japanese).  
Executed Small Entity Status Form.  
Claim of Priority.

09/926826

531 Rec'd

PCT/AT/04/004  
P21844

27 DEC 2004

X. The following fees are submitted:

Basic National Fee (37 CFR 1.492(a)(1)-(5)):

Search report has been prepared by the EPO or JPO. . . . . \$ 890.00

International preliminary examination fee paid to USPTO (37 CFR 1.482). . . . . \$ 710.00

No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO(37 CFR 1.445(a)(2)). . . . . \$ 740.00

Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO. . . . . \$1,040.00

International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4). . . . . \$ 100.00

ENTER APPROPRIATE BASIC FEE AMOUNT =

\$890.00

urcharge of \$130.00 for furnishing the oath or declaration later than \_\_\_\_ 20 \_\_\_\_ 30  
onths from the earliest claimed priority date (37 CFR 1.492(e)).

\$

Claims	Number Filed	Number Extra	RATE		
total Claims	14 - 20 =	0	X \$18.00	\$0.00	
dependent Claims	1 - 3 =	0	X \$84.00	\$0.00	
multiple dependent claim(s) (if applicable)			+ \$280.00	\$0.00	

TOTAL OF ABOVE CALCULATIONS =

\$890.00

- Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced  
by 1/2.

\$445.00

SUBTOTAL =

\$445.00

cessing fee of \$130.00 for furnishing the English translation later than \_\_\_\_ 20 \_\_\_\_ 30  
onths from the earliest claimed priority date (37 CFR 1.492(f)).

+

ention of Time fee in the amount of \$

TOTAL NATIONAL FEE =

\$445.00

e for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be  
ompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property

+

\$40.00

TOTAL FEES ENCLOSED =

\$485.00

Amount to be  
refunded

\$

Charged

\$

X. A check in the amount of \$485.00 to cover the above fees is enclosed.

--- Please charge my Deposit Account No. \_\_\_\_ in the amount of \$ \_\_\_\_ to cover the above fees.

X. The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to  
Deposit Account No. 19-0089.

TE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and  
rested to restore the application to pending status.

ND ALL CORRESPONDENCE TO CUSTOMER NO. 7055

ce H. Bernstein  
THE PRESENT ADDRESS OF:  
ERNELUM & BERNSTEIN, P.L.C.  
1) Roland Clarke Place  
ston, VA 20191  
3) 716-1191



07055

PATENT TRADEMARK OFFICE

*Leslie M. Bernstein*  
SIGNATURE  
Bruce H. Bernstein  
NAME

29,027

REGISTRATION NUMBER

GREENBLUM & BERNSTEIN, P.L.C.  
INTELLECTUAL PROPERTY CAUSES  
1941 ROLAND CLARKE PLACE  
RESTON, VA 20191

Applicant or Patentee: DAIICHI CHEMICAL INDUSTRY CO., LTD. Attorney's \_\_\_\_\_  
Serial or Patent No: \_\_\_\_\_ Docket No.: \_\_\_\_\_  
Filed or Issued: \_\_\_\_\_  
For: \_\_\_\_\_

VERIFIED STATEMENT (DECLARATION) CLAIMING SMALL ENTITY  
STATUS (37 CFR 1.9 (f) and 1.27(c)) - SMALL BUSINESS CONCERN

I hereby declare that I am

☐ the owner of the small business concern identified below:  
☒ an official of the small business concern empowered to act on behalf of the concern identified below:  
NAME OF SMALL BUSINESS CONCERN DAIICHI CHEMICAL INDUSTRY CO., LTD.  
ADDRESS OF SMALL BUSINESS CONCERN Hiranomachi Yachiyo Bldg., 8-13, Hiranomachi  
1-chome, Chujo-ku, Osaka-shi, Osaka 541-0046 JAPAN

I hereby declare that the above identified small business concern qualifies as a small business concern as defined in 13 CFR 121.12, and reproduced in 37 CFR 1.9(d), for purposes of paying reduced fees to the United States Patent and Trademark Office, in that the number of employees of the concern, including those of its affiliates, does not exceed 500 persons. For purposes of this statement, (1) the number of employees of the business concern is the average over the previous fiscal year of the concern of the persons employed on a full-time, part-time or temporary basis during each of the pay periods of the fiscal year, and (2) concerns are affiliates of each other when either, directly or indirectly, one concern controls or has the power to control the other, or a third party or parties controls or has the power to control both.

I hereby declare that rights under contract or law have been conveyed to and remain with the small business concern identified above with regard to the invention, entitled "Flame Retardants for Resins and Flame" by inventor(s) Noriaki TOKIYASU and Katsumi KAMEI, described in Retarded Resin Compositions Containing the Same

☒ the specification filed herewith  
☐ application serial no. \_\_\_\_\_, filed \_\_\_\_\_  
☐ patent no. \_\_\_\_\_, issued \_\_\_\_\_

If the rights held by the above identified small business concern are not exclusive, each individual, concern or organization having rights in the invention is listed below\* and no rights to the invention are held by any person, other than the inventor, who would not qualify as an independent inventor under 37 CFR 1.9(e) if that person made the invention, or by any concern which would not qualify as a small business concern under 37 CFR 1.9(d), or a nonprofit organization under 37 CFR 1.9(e) \*NOTE: Separate verified statements are required from each named person, concern or organization having rights to the invention averring to their status as small entities. (37 CFR 1.27)

FULL NAME \_\_\_\_\_  
ADDRESS \_\_\_\_\_  
☐ INDIVIDUAL ☐ SMALL BUSINESS CONCERN ☐ NONPROFIT ORGANIZATION

FULL NAME \_\_\_\_\_  
ADDRESS \_\_\_\_\_  
☐ INDIVIDUAL ☐ SMALL BUSINESS CONCERN ☐ NONPROFIT ORGANIZATION

I acknowledge the duty to file, in this application or patent, notification of any change in status resulting in loss of entitlement to small entity status prior to paying, or at the time of paying, the earliest of the issue fee or any maintenance fee due after the date on which status as a small entity is no longer appropriate. (37 CFR 1.28 (b))

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application, any patent issuing thereon, or any patent to which this verified statement is directed.

NAME OF PERSON SIGNING Hitoshi YOSHIKAWA  
TITLE OF PERSON IF OTHER THAN OWNER President  
ADDRESS OF PERSON SIGNING 2-1220-26, Nanyama, Nara-shi, Nara 631-0056 JAPAN  
SIGNATURE \_\_\_\_\_ DATE 12/12/2001

Hitoshi YOSHIKAWA

*H. Yoshikawa*

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant : Noriaki TOKUYASU et al.

Serial No : Not Yet Assigned (National Stage of PCT/JP00/04004)

Filed : Concurrently Herewith (International Filing Date June 19, 2000)

For : FLAME RETARDANTS FOR RESINS AND FLAME-RETARDED RESIN  
COMPOSITIONS CONTAINING THE SAME

**PRELIMINARY AMENDMENT**

Commissioner of Patents and Trademarks  
Washington, D.C. 20231

Sir:

Prior to calculation of the filing fees and the examination of the above-identified patent application on the merits, the Examiner is respectfully requested to amend the claims as follows:

**IN THE CLAIMS**

Please amend the claims as follows (a marked-up copy of the claim amendments is provided as an attachment to this Amendment):

3. (Amended-Clean Text) A flame retardant for resins as set forth in claim 1, wherein  $R_3$  in the formula (I) is a straight-chain or branched alkyl group having a carbon number of 1 to 4.

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5. (Amended-Clean Text) A flame-retarded resin composition comprising a resin and a flame retardant for resins as set forth in claim 1.

8. (Amended-Clean Text) A flame-retarded resin composition as set forth in claim 5, wherein 0.1 to 50 parts by weight of the flame-retardant is blended therein with respect to 100 parts by weight of the resin.

Please add new claims 9 to 14 as follows:

---9. A flame retardant for resins as set forth in claim 2, wherein  $R_3$  in the formula (I) is a straight-chain or branched alkyl group having a carbon number of 1 to 4.

10. A flame-retarded resin composition comprising a resin and a flame retardant for resins as set forth in claim 2.

11. A flame-retarded resin composition as set forth in claim 10, wherein the resin does not contain a halogen.

12. A flame-retarded resin composition as set forth in claim 11, wherein the resin is polyurethane.

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13. A flame-retarded resin composition as set forth in claim 6, wherein 0.1 to 50 parts by weight of the flame-retardant is blended therein with respect to 100 parts by weight of the resin.

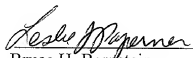
14. A flame-retarded resin composition as set forth in claim 7, wherein 0.1 to 50 parts by weight of the flame-retardant is blended therein with respect to 100 parts by weight of the resin.---

REMARKS

By the above amendment, claims 3, 5, and 8 have been amended and claims 9 to 14 have been added to delete multiple dependency.

If there should be any questions, the Examiner is invited to contact the undersigned at the telephone number listed below.

Respectfully submitted,  
Noriaki TOKUYASU et al.

 Reg. No. 33,329  
Bruce H. Bernstein  
Reg. No. 29,027

December 26, 2001  
GREENBLUM & BERNSTEIN, P.L.C.  
1941 Roland Clarke Place  
Reston, VA 20191  
(703) 716-1191

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MARKED-UP COPY OF AMENDED CLAIMS

3. (Amended) A flame retardant for resins as set forth in claim 1 [or 2], wherein  $R_3$  in the formula (I) is a straight-chain or branched alkyl group having a carbon number of 1 to

4.

5. (Amended) A flame-retarded resin composition comprising a resin and a flame retardant for resins as set forth in claim 1 [or 2].

8. (Amended) A flame-retarded resin composition as set forth in claim 5 [any one of claims 5 to 7], wherein 0.1 to 50 parts by weight of the flame-retardant is blended therein with respect to 100 parts by weight of the resin.

## DESCRIPTION

Flame Retardants for Resins andFlame-Retarded Resin Compositions Containing the Same

## 5 Technical Field

The present invention relates to a flame retardant for resins containing as a main component a phosphoric ester having a phosphorinane structure, and a flame-retarded resin composition containing the ester.

10

## Background Art

Thermoplastic resins such as polypropylene, polystyrene or acrylonitrile-butadiene-styrene (ABS) resin, etc. and thermosetting resins such as polyurethane or phenolic resin, etc. can be produced at relatively low costs and have excellent characteristics such as capability of being easily molded. For this reason, these resins are generally used for a wide variety of everyday articles including electronic components and automobile components.

20

Since the resins themselves are easy to flame, the resins easily burn out once the resins catch fire. Fires especially at public facilities such as electric or communications cables may have serious effects on social functions. Today, flame-retardation is legally demanded with regard to some fields

25

where resins are used such as fields of electric appliances,



automobile interiors, textile products, etc. As such flame-retardation regulations, are known the UL standards for electric appliances in the United State, FMVSS-302 regarding automobiles and the like.

5 In order to provide flame retardant properties to resins, the process of adding flame retardants for resins (referred to as flame retardants hereinafter) during preparation of resin compositions for molding is generally adopted. Such flame retardants include inorganic compounds, organic phosphorus  
10 compounds, organic halogen compounds, halogen-containing organic phosphorus compounds and the like.

Of the above-mentioned compounds, the organic halogen compounds and halogen-containing organic phosphorus compounds exhibit an excellent flame retardant  
15 effect. However, these compounds containing halogen generate hydrogen halide by thermal decomposition during the molding of resins. That will cause problems such as corrosion of mold dies, deterioration of resins and coloring of resins. Further, hydrogen halide worsens work environment because of its  
20 toxicity. In addition to that, the halogen-containing compounds adversely affect human bodies by generating toxic gases such as hydrogen halide and dioxins during burning, for example, in fires.

It is generally considered that coloring which takes  
25 place in the molding of resins is caused by phosphoric acid or

hydrogen chloride generated by dehalogenation reaction and an amine-hydrochloride generated in the co-presence of an amine.

Flame retardants not containing halogens include inorganic compounds such as magnesium hydroxide, aluminum hydroxide, calcium hydroxide, etc. However, these inorganic compounds have only significantly poor flame retardant effects. Consequently, it is necessary to add these compounds in a large amount for obtaining a sufficient flame retardant effect. Thereby, intrinsic properties of resins, especially mechanical properties of molded articles of resins may be impaired.

Therefore, is strongly demanded the development of a flame retardant which does not contain any halogen and is free of the above-described drawbacks.

U.S. Patent No. 5,750,601 discloses 5,5-dimethyl-2-oxo-2-phenoxy-1,3,2-dioxaphosphorinane as a flame retardant not containing a halogen.

However, this compound is solid within the range of room temperature to temperatures at which resins are molded. Therefore, it is difficult to mix the compound with resin materials or to disperse the compound uniformly into the resin materials. For example, a polyurethane foam is obtained by condensation reaction of a diisocyanate with a polyol in which a solid flame retardant is dispersed beforehand. A problem lies in that the flame retardant settles and does not uniformly disperse in the polyol. Also, since the flame retardant is solid,

it is difficult to add the flame retardant in a constant amount automatically and handle the flame retardant when the flame retardant is added to a resin material during the preparation of a resin composition for molding.

5

#### Disclosure of Invention

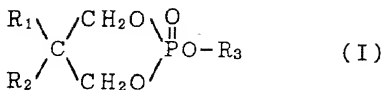
The present invention is to solve the drawbacks of the prior-art techniques as described above, and an object of the present invention is to provide a flame retardant not containing  
10 a halogen which, when added to various kinds of resins, gives excellent flame retardancy to the resins without declining the intrinsic properties of the resins and which has a good resistance to hydrolysis, and a flame-retarded resin composition having excellent mechanical properties.

15 As a result of intensive study to solve the above problems, the inventors have found the blending of a specific phosphoric ester with a resin, and have accomplished the present invention.

Of the phosphoric esters represented by the formula (I)  
20 of the present invention, a compound having methyl groups as  $R_1$  and  $R_2$  and a methyl group as  $R_3$  is known, for example, as a lubricating oil component, but has been found to have an unexpectedly excellent effect when used as a flame retardant.

Accordingly, the present invention provides a flame  
25 retardant for resins containing a phosphoric ester (referred to as

"phosphoric ester (I)" hereinafter) represented by the formula (I):



wherein R<sub>1</sub> and R<sub>2</sub>, the same or different, are an alkyl group having a carbon number of 1 to 6 and R<sub>3</sub> is an alkyl group having a carbon number of 1 to 20.

Also, according to the present invention, there is provided a flame-retarded resin composition (referred to as "a resin composition" hereinafter) containing the above-mentioned flame retardant as a flame retardant in a resin.

#### Best Mode for Carrying Out the Invention

The phosphoric ester contained in the flame retardant of the present invention is represented by the aforesaid formula (I).

In the formula (I), as alkyl groups of R<sub>1</sub> and R<sub>2</sub> having a carbon number of 1 to 6, straight-chain or branched alkyl groups may be mentioned, examples of which include straight-chain alkyl groups such as methyl, ethyl, n-propyl, n-butyl, n-pentyl, n-hexyl, etc., and branched alkyl groups such as isopropyl, isobutyl, sec-butyl, tert-butyl, isopentyl, tert-pentyl, neopentyl, isohexyl, etc. Among these groups, straight-chain or branched alkyl groups having a carbon number of 1 to 4 are preferable, and methyl is the most preferable.

As alkyl groups of  $R_3$  having a carbon number of 1 to 20, straight-chain or branched alkyl groups may be mentioned, among which alkyl groups having a carbon number of 1 to 18 are preferable. Examples thereof include straight-chain alkyl

- 5 groups such as n-heptyl, n-octyl, n-nonyl, n-decyl, n-dodecyl, n-tetradecyl, n-hexadecyl, n-octadecyl, etc., and branched alkyl groups such as methylhexyl, methylheptyl, 2-ethylhexyl, iso-decyl, iso-dodecyl, iso-tetradecyl, iso-hexadecyl, iso-octadecyl, etc., as well as the alkyl groups mentioned above as examples of
- 10  $R_1$  and  $R_2$ . Among these groups, straight-chain or branched lower alkyl groups having a carbon number of 1 to 4 are preferable, and n-propyl and n-butyl are the most preferable.

- Phosphoric esters (I) wherein  $R_3$  is a lower alkyl group are liquid at ordinary temperature and have a low viscosity (15
- 15 to 40 cps/25 °C). They have suitable properties for flame retardants.

As examples of the phosphoric esters (I), the compounds described in Production Examples 1 and 2 may be mentioned.

- 20 The phosphoric esters (I) of the present invention might contain impurities derived from by-products and unreacted materials during production, but may be used as flame retardants without being further purified so long as the impurities do not affect the thermal resistance and the flame
- 25 retardancy of resin compositions.

The flame retardant of the present invention may be a mixture of two or more species of phosphoric esters (I).

The resin composition of the present invention contains a resin and the above-described flame retardant.

5 As examples of the resin, may be mentioned thermoplastic resins such as chlorinated polyethylene, polyethylene, polypropylene, polybutadiene, styrene-based resin, impact-resistant polystyrene, polyvinyl chloride, acrylonitrile - chlorinated polystyrene - styrene (ACS) resin, acrylonitrile -  
10 styrene (AS) resin, ABS resin, polyphenylene ether, modified polyphenylene ether, polymethacrylate, polyamide, polyester, polycarbonate, etc., and thermosetting resins such as polyurethane, phenolic resin, epoxy resin, melamine resin, urea resin, unsaturated polyester, etc. These resins may be used as  
15 a mixture of two or more thereof.

Among the above-mentioned resins, those not containing a halogen may be preferable.

In other words, it is possible to cope perfectly with the generation of halogen-containing toxic gases which may have  
20 various adverse effects by combining the flame retardant which does not contain a halogen with a resin which does not contain a halogen as well.

Polyurethane is particularly preferable among the resins not containing a halogen.

25 Polyurethane is produced by condensation reaction of a

polyol with a diisocyanate. A flame retardant is used by dispersing it in a polyol. Since the flame retardant of the present invention is a liquid of low viscosity, the flame retardant can be uniformly dispersed in the polyol and furthermore can  
5 reduce the viscosity of the polyol, which is highly viscous intrinsically, thereby eliminating problems of sedimentation and dispersion failure.

Further, the flame retardant of the present invention can be automatically added at a constant addition rate to a resin  
10 material or during preparation of a resin composition for molding. A pre-mixture of the flame retardant and the polyol can be stored. Therefore, workability and storability are improved.

Since the flame retardant is uniformly blended in the  
15 obtained polyurethane, excellent flame retardancy is maintained for a long time.

The amount of the flame retardant blended in the resin composition of the present invention may be selected as appropriate depending upon the kind of a phosphoric ester (I),  
20 the kind of a resin, the use of a molded article of the resin composition and the performance (e.g., flame retardancy, etc.) required of the molded article.

The amount of the phosphoric ester (I) blended as a flame retardant may typically be 0.05 parts by weight or more,  
25 preferably 0.1 to 50 parts by weight, more preferably 5 to 20

parts by weight, with respect to 100 parts by weight of the resin. If the blend amount of the phosphoric ester (I) is less than 0.05 parts by weight, it is not preferable because the resin cannot be provided with sufficient flame retardancy. If the blend amount  
5 of the phosphoric ester (I) is more than 50 parts by weight, it is not preferable because it may cause a decline in properties of the resin. In the case where the resin is polyurethane, the flame retardant may preferably be added in the step of producing polyurethane. As regards the blend amount of the flame  
10 retardant in this case, the phosphoric ester (I) may be blended in an amount within the above-mentioned range with respect to 100 parts by weight of the total amount of a polyol and a diisocyanate, which are starting materials.

Other additives for resins may be blended, if necessary,  
15 in the resin composition of the present invention so long as the blending of the additives does not impair the properties of the resin to which the flame retardancy should be given.

As such additives for resins, may be mentioned other flame retardants than the phosphoric ester (I), antioxidants,  
20 inorganic fillers, antistatics, ultraviolet absorbers, lubricants and the like.

As the flame retardants other than the phosphoric ester (I), may be mentioned organic phosphorus compounds such as trimethyl phosphate, triethyl phosphate, tributyl phosphate,  
25 triphenyl phosphate, tricresyl phosphate, cresyldiphenyl



phosphate, tri-2-ethylhexyl phosphate, etc.; nitrogen-containing compounds such as melamine, benzoguanamine, urea, ammonium polyphosphate and ammonium pyrophosphate; metal compounds such as aluminum hydroxide, magnesium hydroxide, zinc borate, etc.; and the like.

As the antioxidants, may be mentioned phosphorus compounds such as trivalent phosphorous compounds including triphenyl phosphite, tris(nonylphenyl)phosphite, diphenylisodecyl phosphite, bis(2,4-di-tert-butylphenyl)pentaerythritol diphosphite, tetrakis(2,4-di-tert-butylphenyl)-4,4-diphenylene phosphonite, etc.; hydroquinone compounds such as hydroquinone, 2,5-di-tert-butylhydroquinone, octylhydroquinone, 2,5-tert-amylhydroquinone, etc.; phenol compounds; amine compounds; sulfur compounds; and the like.

As the inorganic fillers, may be mentioned mica, talc, alumina and the like.

As the antistatics, may be mentioned cationic surfactants, non-ionic surfactants and the like.

As the ultraviolet absorbers, may be mentioned benzophenone compounds, salicylate compounds, benzotriazole compounds and the like.

As the lubricants, may be mentioned fatty acid compounds, fatty amide compounds, ester compounds, alcohol compounds and the like.

For producing the resin composition of the present invention, there is no particular limitation on the order or method of blending the respective components.

For example, the resin composition may be produced by  
5 mixing, melting and kneading the flame retardant, the resin and other additives for resin, as required, using known methods. For mixing, melting and kneading, general-use apparatus such as a single-screw extruder, a double-screw extruder, a Banbury mixer, a kneader mixer and the like may be used singly or in  
10 combination.

If the resin is produced by bulk polymerization, the flame retardant may be added ① at the feeding of a monomer, ② at the last stage of bulk polymerization reaction or ③ at the molding of the resulting polymer.

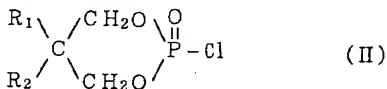
15 The obtained resin composition can be further molded by a known method to obtain a molded product in a desired form, for example, a plate, sheet or film form.

The phosphoric ester (I) of the present invention may be produced, for example, by the method disclosed by U.S. Patent  
20 No. 5,750,601 (however, a saturated aliphatic alcohol is used in place of phenol).

Now explanation is given of methods of producing the phosphoric ester (I) for reference.  
(Production Method 1)

25 Phosphorus oxychloride is reacted with 0.9 to 1.2 mol

(preferably, 0.95 to 1.05 mol) of a diol compound with respect to 1 mol of phosphorus oxychloride, to obtain a compound represented by the formula (II):



5 (wherein  $\text{R}_1$  and  $\text{R}_2$ , the same or different, are an alkyl group having a carbon number of 1 to 6).

As the diol compound, a 2,2-dialkyl-1,3-propanediol is preferable. Examples thereof include 2,2-dimethyl-1,3-propanediol (= neopentyl glycol), 2-methyl-2-ethyl-1,3-propanediol, 2,2-diethyl-1,3-propanediol, 2,2-di-n-butyl-1,3-propanediol, 2,2-di-sec-butyl-1,3-propanediol, 2,2-di-tert-butyl-1,3-propanediol, 2-ethyl-2-n-butyl-1,3-propanediol, 2,2-di-n-pentyl-1,3-propanediol, 2,2-di-n-octyl-1,3-propanediol and the like, of which neopentyl glycol is the most preferable from  
10 the viewpoint of price and availability.

Since the produced compound (II) is solid, this reaction may preferably be carried out in an organic solvent in order to progress the reaction smoothly and avoid difficulty in handling the product owing to crystallization of the product.

20 Preferably, the organic solvent is inert, that is, the organic solvent does not involve a side reaction with phosphorus oxychloride, the diol compound and hydrogen chloride which is by-produced during the reaction.

Particularly, are preferable hydrocarbon solvents such as hexane, cyclohexane, heptane, octane, benzene, toluene, xylene, petroleum spirit, etc.; halogen-containing hydrocarbon solvents such as chloroform, carbon tetrachloride, 1,2-dichloroethane, trichloroethane, tetrachloroethane, chlorobenzene, dichlorobenzene, etc.; ether solvents such as diisopropyl ether, dibutyl ether, 1,4-dioxane, ethylene glycol diethyl ether, etc.; and the like, among which toluene, 1,2-dichloroethane and 1,4-dioxane are preferable, and 1,4-dioxane is particularly preferable.

The amount of the organic solvent used is not particularly limited, but may preferably be about 35 to 70 wt% with respect to the diol compound.

The reaction may preferably be carried out by stirring a mixture of the diol compound and the organic solvent and adding phosphorus oxychloride to the mixture.

The reaction temperature is 0 to 100 °C, preferably 40 to 80 °C. If the reaction temperature is lower than 0 °C, it is not preferable because the reaction does not progress sufficiently. If the reaction temperature exceeds 100 °C, it is not preferable because the produced product (II) decomposes and colors.

The reaction time is typically about 3 to 8 hours, though it may be varied depending upon conditions such as the reaction temperature.

By-produced hydrogen chloride can be removed by a known method, for example, a vacuum treatment such as nitrogen gas topping. The conditions of the vacuum treatment are preferably at such degrees that the organic solvent is not  
5 refluxed, and, for example, are a pressure within the range of 200 to 500 Torr at 40 to 90 °C, preferably 50 to 80 °C.

Subsequently, the compound (II) is reacted with a theoretical amount to a 20 wt% excess of the theoretical amount (preferably, an amount within the range of 10 to 15 wt% excess  
10 of the theoretical amount) of an alcohol to obtain the phosphoric ester (I).

The theoretical amount of the alcohol is calculated by the following formula:

The theoretical amount of the alcohol  
15 
$$= (A \times B \times C) / (35.5 \times 100)$$

(wherein A is the weight (g) of the compound (II), B is the chlorine content (wt%) in the compound (II) and C is the molecular weight of the alcohol).

If the amount of the alcohol used is less than the  
20 theoretical amount, the reaction is not completed. If the amount of the alcohol used exceeds 20 wt%, it is not economically preferable because an unreacted alcohol remains in a reaction system.

As the alcohol, may be mentioned methanol, ethanol,  
25 1-propanol, 2-propanol, 1-butanol, sec-butanol, tert-butanol,

hexanol, heptanol and octanol, among which 1-propanol and 1-butanol are particularly preferable.

This reaction may preferably be carried out in the presence of an acid-acceptor (for example, a base such as triethylamine, tributylamine or the like) for the purpose of reducing the reaction time and improving the purity of the product.

The amount of the acid-acceptor used is within the range of a theoretical amount to a 20 wt% excess of the theoretical amount, preferably within the range of 10 to 15 wt% excess of the theoretical amount.

The theoretical amount of the acid-acceptor is calculated by the following formula:

The theoretical amount of the acid-acceptor

$$= (A \times B \times D) / (35.5 \times 100)$$

(wherein A is the weight (g) of the compound (II), B is the chlorine content (wt%) in the compound (II) and D is the molecular weight of the acid-acceptor).

This reaction can be carried out continuously to the previous step reaction, particularly, by adding the alcohol or a mixture of the alcohol and the acid-acceptor while stirring the reaction product of the previous step containing the organic solvent.

The reaction temperature is 20 to 60 °C, preferably 30 to 50 °C. If the reaction temperature is lower than 20 °C, it is

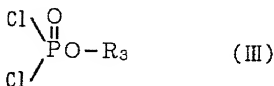
not preferable because the reaction progresses very slowly. If the reaction temperature exceeds 60 °C, it is not preferable because a side reaction may take place to cause adverse effects such as a decline in the purity of the product.

5           The reaction time is typically about 2 to 7 hours, though it may be varied depending upon conditions such as the reaction time.

Hydrogen chloride by-produced through this reaction is taken by the acid-acceptor to form a salt. This salt can be  
10 removed by washing the reaction mixture after the reaction is completed. More particularly, the obtained reaction mixture is collected from a reaction vessel and subjected to purification treatment such as washing and dehydration. For removing remaining water and low-boiling contents, it is preferable to  
15 carry out steam distillation as purification treatment.

(Production Method 2)

First, an alcohol is reacted with 1.0 to 5.0 mol (preferably, 1.1 to 3.0 mol) of phosphorus oxychloride with respect to 1 mol of the alcohol to obtain a compound represented  
20 by the formula (III):



(wherein R<sub>3</sub> is an alkyl group having a carbon number of 1 to 20).

As the alcohol, may be mentioned the alcohols mentioned in the above described Production Method 1.

The reaction temperature is - 20 to 50 °C, preferably - 10 to 20 °C. If the reaction temperature is lower than -20 °C, it is not preferable because the reaction does not progress sufficiently. If the reaction temperature exceeds 50 °C, it is not preferable because the by-produced hydrogen chloride reacts with the alcohol to generate an alkyl chloride and water, which in turn decomposes the compound (III) and phosphorus oxychloride.

The reaction time is typically about 3 to 8 hours, though it may be varied depending upon conditions such as the reaction temperature.

By-produced hydrogen chloride can be removed by a known method, for example, a vacuum treatment such as nitrogen gas topping. The conditions of the vacuum treatment are, for example, a pressure within the range of 10 to 200 Torr at 0 to 20 °C, preferably 5 to 10 °C.

Since the compound (III) decomposes at about 50 °C, the above-mentioned temperature conditions are preferably set at or below 50 °C.

Unreacted phosphorus oxychloride remaining in a system is preferably removed after the reaction is completed because there are problems, for example, in that the unreacted phosphorus oxychloride will cause a side reaction with a diol



compound in the next step reaction to decline the purity of the product.

Next, the compound (III) is reacted with a theoretical amount to a 20 wt% excess of the theoretical amount (preferably  
5 amount within the range of 10 to 15 wt% excess of the theoretical amount) of the diol compound to obtain the phosphoric ester (I).

The theoretical amount of the diol compound is calculated by the following formula:

10 The theoretical amount of the diol compound

$$= (E \times F \times G) / [2 \times (35.5 \times 100)]$$

(wherein E is the weight (g) of the compound (III), F is the chlorine content (wt%) in the compound (III) and G is the molecular weight of the diol compound).

15 Since the diol compound is solid, this reaction may preferably be carried out in an organic solvent in order to progress the reaction smoothly and avoid difficulty in handling the product owing to crystallization of the product.

As the diol compound and the organic solvent, may be  
20 mentioned those mentioned in the above-described Production Method 1. The amount of the organic solvent used is not particularly limited, by may preferably be 35 to 70 wt% with respect to the diol compound.

This reaction may preferably be carried out in the  
25 presence of an acid-acceptor similar to those of Production

Method 1 for the purpose of reducing the reaction time and improving the purity of the product.

The amount of the acid-acceptor used is within the range of a theoretical amount to a 20 wt% excess of the theoretical amount, preferably within the range of 10 to 15 wt% excess of the theoretical amount.

The theoretical amount of the acid-acceptor is calculated by the following formula:

$$\text{The theoretical amount of the acid-acceptor} \\ = (E \times F \times C) / (35.5 \times 100)$$

(wherein E is the weight (g) of the compound (III), F is the chlorine content (wt%) in the compound (III) and C is the molecular weight of the alcohol).

The reaction temperature is 20 to 60 °C, preferably 30 to 50 °C. If the reaction temperature is lower than 20 °C, it is not preferable because the reaction progresses very slowly. If the reaction temperature exceeds 60 °C, it is not preferable because a side reaction may take place to cause adverse effects such as a decline in the purity of the product.

The reaction time is typically about 2 to 7 hours, though it may be varied depending upon conditions such as the reaction temperature.

Hydrogen chloride by-produced through this reaction is taken by the acid-acceptor to form a salt. This salt can be removed by washing the reaction mixture after the reaction is

completed. More particularly, the obtained reaction mixture is collected from a reaction vessel and subjected to purification treatment such as washing and dehydration. For removing remaining water and low-boiling contents, it is preferable to carry out steam distillation as purification treatment.

### Examples

The present invention is now explained in further detail with reference to production examples and examples, which however are not intended to limit the scope of the invention. In the examples, "a part or parts" means "a part or parts by weight" unless otherwise indicated.

#### Production Example 1

In a four-necked 1-L flask provided with a stirrer, a thermometer, a dropping funnel and a condenser connected to a water scrubber, 204.0 g (2.0 mol) of neopentyl glycol and 102.0 g of 1,4-dioxane (50 wt% with respect to neopentyl glycol) were fed and stirred. Subsequently, the resulting mixture was heated to 50 °C using a thermostatic device. With maintaining this temperature, 307.0 g (2.0 mol) of phosphorus oxychloride was added in three hours through the dropping funnel. After addition, the reaction mixture was stirred at 50 °C for two hours, and further the reaction mixture was heated to 75 °C in an hour. Subsequently, by-produced hydrogen chloride was removed at a

vacuum degree of 250 Torr at 75 °C in two hours, to obtain 471.0 g of Compound (II) wherein R<sub>1</sub> and R<sub>2</sub> were methyl. The chlorine content in this compound was 15.07 %.

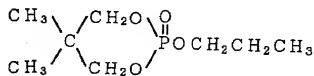
Subsequently, the above reaction mixture was cooled to 50 °C. With maintaining this temperature, 354.1 g of a mixed solution of 222.1 g (2.2 mol) of triethylamine and 132.0 g (2.2 mol) of 1-propanol were added to the reaction mixture in two hours. After addition, the reaction mixture was stirred for three hours with maintaining its temperature at 50 °C.

The obtained reaction mixture was washed with water to remove triethylamine hydrochloride. Further water and low-boiling contents were removed by heating under vacuum, to give 395.2 g of a pale yellow liquid.

The obtained product was elementally analyzed and its viscosity was measured.

The obtained product was a Compound (1) having the following chemical structure.

Table 1 shows the results of elemental analysis and theoretical values, the viscosity and the yield of the product.



Compound (1)

#### Production Example 2

A pale yellow liquid, 421.8 g, was obtained in the same

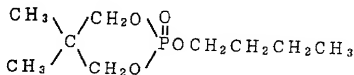
manner as described in Production Example 1 except that 162.8 g (2.2 mol) of 1-buthanol were used in place of 1-propanol.

The obtained product was elementally analyzed and its viscosity was measured in the same manner as described in

#### 5 Production Example 1.

The product is a Compound (2) having the following chemical structure.

Table 1 shows the results of elemental analysis and theoretical values, the viscosity and the yield of the product.



10

Compound (2)

Table 1

	Composition			Viscosity (cps/25℃)	Yield (%)
	Upper : Analytic results				
	Lower : Theoretical values				
	P %	C %	H %		
Compound (1)	14.88	46.14	8.16	21	95.0
	14.90	46.15	8.17		
Compound (2)	13.94	48.63	8.54	26	95.0
	13.96	48.65	8.56		

15

In the following examples, performance evaluation was carried out on Compound (1) and Compound (2), which were flame retardants in accordance with the present invention, as

well as a Compound (A) and a Compound (B), which were known flame retardants not containing a halogen as comparative examples.

Compound (A) : triethyl phosphate

5       Compound (B) : cresyldiphenyl phosphate

Example 1 (Evaluation of flame retardants on resistance to hydrolysis)

Compound (1) and Compound (2), where were obtained  
10 in Production Example 1 and Production Example 2, respectively, Compound (A) and Compound (B) were measured for their acid values as evaluation of resistance to hydrolysis in accordance with ASTM D-2169 and MIL III-19457.

In a pressure-resistant sample bottle, 75 g of each  
15 compound to be measured and 25 g of distilled water were fed as samples, and the bottle was capped tightly. Then the pressure-resistant sample bottle was attached to a hydrolyzing apparatus (which has the function of rotating five times in a minute to mix the contents of a sample bottle) adjusted to 93 °C  
20 beforehand, maintained at the same temperature for 48 hours and cooled to room temperature.

The mixture in the pressure-resistant sample bottle was moved into a separatory funnel and allowed to stand, and an aqueous phase was collected. Subsequently, about 100 g of  
25 distilled water were added as washing water to an oily phase,



	Organic Chemicals Co., Ltd.)	0.3 parts
	Amine-based catalyst (Tradename : Kaolizer No. 1, produced by Kao Corporation)	0.1 parts
	Water	5.0 parts
5	Dichloromethane	5.0 parts

Each of the flame retardants (in an amount with respect to 100 parts by weight of the total amount of the polyol and diisocyanate, shown in Table 3)

- With use of the above-mentioned components, flexible
- 10 urethane foams were produced by a one-shot process as described below.

First, the polyol, silicone oil, catalysts, dichloromethane, water and flame retardant were blended and uniformly mixed by stirring for a minute by a stirrer at a

15 revolution rate of 3000 rpm. Subsequently, the diisocyanate was added and the resulting mixture was further stirred for 5 to 7 seconds at a revolution rate of 3000 rpm. Subsequently the resulting mixture was poured quickly into a board-carton box of square cross section. The mixture started foaming immediately

20 and reached the maximum volume after several minutes. This foamed product was cured for 30 minutes in an oven at 80 °C. The resulting foamed product had a cellular structure of white flexible foam type.

- 25 Example 3 (Production of flame-retarded rigid polyurethane



foams)

Polyol (Tradename : SU-464, produced by Mitsui Chemicals, Inc.) 100 parts

Diisocyanate (Tradename : M-200, produced by Mitsui Chemicals, Inc.) 192.2 parts

Silicone oil (Tradename : SH-193, produced by Dow Corning Toray Silicone Co., Ltd.) 2.0 parts

Tin-based catalyst (Tradename : STANN BL, produced by Sankyo Organic Chemicals Co., Ltd.) 0.1 parts

Amine-based catalyst (Tradename : Kaolizer No. 3, produced by Kao Corporation) 0.7 parts

Water 5.0 parts

Each of the flame retardants (in an amount with respect to 100 parts by weight of the total amount of the polyol and

diisocyanate, shown in Table 4)

With use of the above-mentioned components, rigid urethane foams were produced by a one-shot process as described below.

First, the polyol, silicone oil, catalysts, water and flame retardant were blended and uniformly mixed by stirring for a minute by a stirrer at a revolution rate of 3000 rpm. Subsequently, the diisocyanate was added and the resulting mixture was further stirred for 5 to 7 seconds at a revolution rate of 3000 rpm. Subsequently, the resulting mixture was poured quickly into a board-carton box of square cross section.

The mixture started foaming immediately and reached the maximum volume after several minutes. The resulting foamed product had a cellular structure of white rigid foam type.

5    Example 4 (Performance evaluation of flame-retarded flexible polyurethane foams)

The flame-retarded flexible polyurethane foams obtained in Example 2 were determined for their properties in accordance with the following standards or by the following

10    operation.

① Density ( $\text{kg/m}^3$ ) : determined in accordance with JIS K-7222

② 25 % hardness (kgf) : determined in accordance with JIS K-6401

15    ③ Flame retardancy : evaluated in accordance with FMVSS-302 (Flame retardancy test for flexible polyurethane foams)

Evaluation criteria :

20    NB : self-extinguished within a marked line A (38 mm from an ignition point)

SE : self-extinguished within a range from the marked line A to a marked line B (38 to 292 mm from the ignition point)

25    BN : burned over the marked line B (292 mm from the ignition point)

#### ④ Susceptibility to discoloration

A test piece ( $5 \times 5 \times 1 \text{ cm}^3$ ) was put in a thermostatic chamber at  $120^\circ \text{C}$  and maintained for 24 hours. Thereafter, the degree of discoloration (YI) of the test piece was determined for indicating the susceptibility to discoloration using a colorimeter.

The obtained results are shown in Table 3

Table 3

Evaluation No.	Flame retardant	Use amount* (parts)	Density ( $\text{kg/m}^3$ )	25% hardness (kgf)	Flame retardancy	Susceptibility to discoloration (YI)
1	Compound (1)	5.6 (9)	25	10	NB	30
2	Compound (2)	6.3 (10)	25	10	NB	30
3	Compound (A)	6.3 (10)	25	10	SE	80
4	Compound (B)	6.3 (10)	25	10	BN	30

\* Blended amount with respect to 100 parts by weight of the total amount of the polyol and diisocyanate (blended amount with respect to 100 parts by weight of the polyol)

Example 5 (Performance evaluation of flame-retarded rigid polyurethane foams)

The flame-retarded rigid polyurethane foams obtained in Example 3 were determined for their properties in accordance

with the following standards or by the following operation.

① Density ( $\text{kg}/\text{m}^3$ ) : determined in accordance with JIS K-7222

② Flexural strength ( $\text{kgf}/\text{cm}^2$ ) : determined in accordance with JIS K-7220

③ Flame retardancy : evaluated in accordance with JIS A-9514 (Testing method for flame retardancy of rigid polyurethane foams)

The obtained results are shown in Table 4

Table 4

Evaluation No.	Flame retardant	Use amount * (parts)	Density ( $\text{kg}/\text{m}^3$ )	Flexural strength ( $\text{kgf}/\text{cm}^2$ )	Flame Retardancy (mm)
5	Compound (1)	3.4 (10)	32	2	36.7
6	Compound (2)	4.1 (12)	32	2	38.6
7	Compound (A)	2.7 (8)	32	2	39.6
8	Compound (B)	4.1 (12)	32	2	Burned out

\* Blended amount with respect to 100 parts by weight of the total amount of the polyol and diisocyanate (blended amount with respect to 100 parts by weight of the polyol)

The results in Table 3 and Table 4 show the following:

① The flame retardants of the present invention, added

to the resin in a smaller amount as compared with the prior-art flame retardants not containing a halogen, can provide excellent flame retardancy to the resins without declining the intrinsic properties of the resins.

- 5           ② The resin compositions (polyurethane foams) of the present invention are excellent in the resistance to discoloration.

Generally, the discoloration (coloration) of resin compositions is considered to be due to compounds generated  
10 when flame retardants themselves hydrolyze. It is therefore considered that the excellent resistance to hydrolysis of the flame retardants of the present invention contributes to suppression of the discoloration of the resin compositions.

- 15           The flame retardancy of the present invention, when mixed with various resins, can provide excellent flame retardancy to the resins without declining the intrinsic properties of the resins. Further, since the flame retardancy of the present invention does not contain a halogen and does not  
20 generate toxic gases during the molding or burning of the resins, the flame retardant does not put loads on the environment or does not affect human bodies adversely.

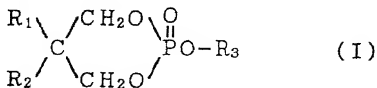
Furthermore, the flame retardant of the present invention is a liquid of low viscosity at room temperature,  
25 disperses well in resins and exhibits good compatibility

especially to polyurethane. In addition, since it is excellent in the resistance to hydrolysis, the flame retardant of the invention does not discolor resin compositions and does not decline durability.

5           The resin composition of the present invention can be suitably used for domestic electric appliances such as VTRs, distribution boards, television sets, air conditioners, etc., OA equipment including personal computers, printers, facsimile machines, telephones, etc., electric devices such as connectors,  
10 switches, motor components, etc., automobile components such as console boxes, seat cushions, etc., construction materials and the like.

# CLAIMS

1. A flame retardant for resins containing a phosphoric ester represented by the formula (I):



5 wherein  $\text{R}_1$  and  $\text{R}_2$ , the same or different, are an alkyl group having a carbon number of 1 to 6 and  $\text{R}_3$  is an alkyl group having a carbon number of 1 to 20.

2. A flame retardant for resins as set forth in claim 1, wherein  $\text{R}_1$  and  $\text{R}_2$  in the formula (I) are straight-chain or  
10 branched alkyl groups having a carbon number of 1 to 4.

3. A flame retardant for resins as set forth in claim 1 or 2, wherein  $\text{R}_3$  in the formula (I) is a straight-chain or branched alkyl group having a carbon number of 1 to 4.

4. A flame retardant for resins as set forth in claim 1,  
15 wherein  $\text{R}_1$  and  $\text{R}_2$  in the formula (I) are methyl groups and  $\text{R}_3$  is a n-propyl or n-butyl group

5. A flame-retarded resin composition comprising a resin and a flame retardant for resins as set forth in claim 1 or 2.

6. A flame-retarded resin composition as set forth in claim  
20 5, wherein the resin does not contain a halogen.

7. A flame-retarded resin composition as set forth in claim 6, wherein the resin is polyurethane.

8. A flame-retarded resin composition as set forth in any

one of claims 5 to 7, wherein 0.1 to 50 parts by weight of the flame-retardant is blended therein with respect to 100 parts by weight of the resin.



# Declaration and Power of Attorney For Utility or Design Patent Application

## 特許出願宣言書

### Japanese Language Declaration

私は、下欄に氏名を記載した発明者として、以下のとおり宣言する：

私の住所、郵便の宛先および国籍は、下欄に氏名に続いて記載したとおりであり、

名称の発明に関し、請求の範囲に記載した特許を求める主題の本来の、最初にして唯一の発明者である（一人の氏名のみが下欄に記載されている場合）か、もしくは本来の、最初にして共同の発明者である（複数の氏名が下欄に記載されている場合）と信じ、

上記発明の明細書（下記の欄でX印がついていない場合は、本書に添付）は、

☐ 年 月 日に提出され、  
米国出願番号 として、  
（該当する場合） 年 月 日に訂正されました。又は、  
特許協定条約国際出願番号 として、  
（該当する場合） 年 月 日に訂正されました。

私は、前記のとおり補正した請求の範囲を含む前記明細書の内容を検討し、理解したことを陳述する。

私は、連邦規則法典第37編第1条第56項に定義されたとおり、特許資格の有無について重要な情報を開示すべき義務があることを認めます。

私は合衆国法典第35部第119条(a-d)項又は第365条(b)項に基づき、下記の外特許出願又は発明者証出願、或いは第365条(a)項に基づき、少なくとも米国以外の1ヶ国を指名したPCT国際出願の外国優先権を主張し、更に優先権の主張に係る基礎出願の出願日前の出願日を有する外国特許出願、又は発明者証出願、或いはPCT国際出願を以下に“なし”の欄に印をつけることにより明記する：

Prior foreign applications  
先の外特許出願

Hei 11(1999)-195058 JAPAN  
(Number) (Country)  
(番号) (国名)

09/07/1999  
(Day/Month/Year Filed)  
(出願の年月日)

(Number) (Country)  
(番号) (国名)

(Day/Month/Year Filed)  
(出願の年月日)

☐ その他の外国特許出願番号は別紙の追補優先権欄に記載する。

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

"Flame Retardants for Resins and Flame-Retarded Resin Compositions Containing the Same"

the specification of which is attached hereto unless the following box is checked:

☐ was filed on \_\_\_\_\_ as  
United States Application Number \_\_\_\_\_  
and was amended on \_\_\_\_\_ (if applicable) or,  
PCT International Application Number \_\_\_\_\_  
and was amended on \_\_\_\_\_ (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, §1.56.

I hereby claim foreign priority under Title 35, United States Code §119(a-d) or §365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT international application which designated at least one country other than the United States, listed below. I have also identified below, by checking the "No" box, any foreign application for patent or inventor's certificate, or of any PCT international application having a filing date before that of the application on which priority is claimed:

Priority claimed  
優先権の主張  
☒ Yes ☐ No  
あり なし  
☐ Yes ☐ No  
あり なし

☐ Additional foreign application numbers are listed on a supplemental priority sheet attached hereto.

# Japanese Language Utility or Design Patent Application Declaration

私は、合衆国法典第35部第119条(c)項に基づく、下記の合衆国仮特許出願の利益を主張する。

I hereby claim the benefit under Title 35, United States Code §119(c) of any United States provisional application(s) listed below.

(Application No.) (出願番号)	(Day/Month/Year Filed) 出願の年月日
(Application No.) (出願番号)	(Day/Month/Year Filed) 出願の年月日
(Application No.) (出願番号)	(Day/Month/Year Filed) 出願の年月日

☐ その他の合衆国仮特許出願番号は別紙の追補優先権欄にて記載する。

☐ Additional provisional application numbers are listed on a supplemental priority sheet attached hereto.

私は、合衆国法典第35部第120条に基づく下記の合衆国特許出願、又は第365条(c)項に基づく合衆国を指名したPCT国際出願の利益を主張し、本願の請求の範囲各項に記載の主題が合衆国法典第35部第112条第1項規定の態様で、先の合衆国特許出願又はPCT国際出願に開示されていない現度において、先の出願の出願日と本願の国内出願日又はPCT国際出願日の間に有効となった連邦規則法典第37部第1章第56条に記載の特許要件に必要の情報を開示すべき義務を有することを認める。

I hereby claim the benefit under Title 35, United States Code §120 of any United States application(s), or §365(c) of any PCT international application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT international application in the manner provided by the first paragraph of Title 35, United States Code §112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

PCT/JE00/04004	19/06/2000	Pending	(Status)
(Application No.) (出願番号)	(Day/Month/Year Filed) 出願の年月日	(現況) (特許済み、係属中 放棄済み)	(patented, pending, abandoned)
(Application No.) (出願番号)	(Day/Month/Year Filed) 出願の年月日	(現況) (特許済み、係属中 放棄済み)	(Status) (patented, pending, abandoned)

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# Japanese Language Utility or Design Patent Application Declaration

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顧客番号 7055

現在選任された弁護士は下記の通りである。

Neil F. Greenblum  
Bruce H. Bernstein  
James L. Rowland  
Arnold Turk

POWER OF ATTORNEY: As a named inventor, I hereby appoint the attorney(s) and/or agent(s) associated with the Customer Number provided below to prosecute this application and transact all business in the Patent and Trademark Office connected therewith, and direct that all correspondence be addressed to that Customer Number:

CUSTOMER NUMBER 7055

The appointed attorneys presently include:

Reg. No. 28,394  
Reg. No. 29,027  
Reg. No. 32,674  
Reg. No. 33,094

Address: GREENBLUM & BERNSTEIN, P.L.C.  
1941 ROLAND CLARKE PLACE  
RESTON, VA 20191

直接電話連絡先: (名称および電話番号)

Direct Telephone Calls to: (name and telephone number)

GREENBLUM & BERNSTEIN, P.L.C.  
(703) 716-1191

唯一のまたは第一の発明者の氏名

同発明者の署名

日付

Full name of sole or first inventor

Noriaki TOKUNASU

Inventor's signature

Date

Noriaki Tokunasu 13.12.2001

住所

Residence

Tokai, JAPAN

国籍

Citizenship

JAPAN

郵便の宛先

Post Office Address

5-420, Koyanwaki, Kagiya-cho, Tokai-shi,

第2の共同発明者の氏名 (該当する場合)

同第2共同発明者の署名

日付

Aichi 477-0032 JAPAN

Full name of second joint inventor, if any

Katsumi KAMEDA

Second inventor's signature

Date

Katsumi Kameda 13.12.2001

住所

Residence

Chita, JAPAN

国籍

Citizenship

JAPAN

郵便の宛先

Post Office Address

10-1, Ishihama Ashima, Higashiura-cho, Chita-gun,

Aichi 470-2103 JAPAN

(第六またはそれ以降の共同発明者に対しても同様な情報および署名を提供すること。)

(Supply similar information and signature for third and subsequent joint inventors.)